

Air quality assessment for Portugal

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Abstract

According to the Air Quality Framework Directive, air pollutant concentration levels have to be assessed and reported annually by each European Union member state, taking into consideration European air quality standards. Plans and programmes should be implemented in zones and agglomerations where pollutant concentrations exceed the limit and target values. The main objective of this study is to perform a long-term air quality simulation for Portugal, using the CHIMERE chemistry-transport model, applied over Portugal, for the year 2001. The model performance was evaluated by comparing its results to air quality data from the regional monitoring networks and to data from a diffusive sampling experimental campaign. The results obtained show a modelling system able to reproduce the pollutant concentrations' temporal evolution and spatial distribution observed at the regional networks of air quality monitoring. As far as the fulfilment of the air quality targets is concerned, there are excessive values for nitrogen and sulfur dioxides, ozone also being a critical gaseous pollutant in what concerns hourly concentrations and AOT40 (Accumulated Over Threshold 40 ppb) values.

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1. Introduction

Air quality is one of the areas in which Europe has been most active in recent years. The European Commission (EC) defined an overall strategy through the setting of long-term air quality objectives. A series of directives has been introduced to control levels of certain pollutants and to monitor their concentrations in the air. In 1996, the Environment Council adopted the Framework Directive 96/62/EC (FWD) on ambient air quality assessment and management. This directive

covers the revision of previously existing legislation and introduces new air quality standards for previously unregulated air pollutants, setting the timetable for the development of daughter directives for a range of pollutants. The list of atmospheric pollutants to be considered includes sulfur dioxide (SO₂), nitrogen dioxide (NO₂), particulate matter, lead, ozone (O₃), and benzene, carbon monoxide (CO), polyaromatic hydrocarbons, and some heavy metals.

The FWD was followed by the daughter directives, which establish the numerical limit values, or, in the case of ozone, target values, for each of the identified pollutants. Besides setting air quality limit and alert thresholds, the objectives of the daughter directives are to harmonize monitoring strategies, measuring methods,

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calibration, and quality assessment methods to arrive at comparable measurements throughout the European Union (EU) and to provide good public information.

In terms of ambient air quality assessment, the FWD also introduces new guidelines, identifying modelling as an air quality management tool and defining that ambient air quality, throughout the territory of the member states, shall be assessed by a combination of measurements and modelling techniques to provide an adequate level of information on ambient air quality in respect to the relevant pollutants. In fact, measured concentrations available at given monitoring sites do not generally describe sufficiently the spatial distribution of air pollutants over wide areas, whereas this information is a crucial factor to evaluate the impact of air pollution on human health and natural ecosystems. Modelling systems can represent suitable tools for these purposes, allowing the study of air quality with an adequate spatial detail, the verification of the fulfilment of the limit targets and threshold values imposed by the EC directives, and also the assessment of appropriate emission reduction strategies.

Therefore, to assess the air quality at a regional level, verifying the fulfilment of the limit targets and threshold values imposed by the EC directives, and to understand the causes and origin of air pollution, numerical modelling exercises should be used. Recent works (Hogrefe et al., 2001) emphasize the importance of policy analysis on a “climatological” basis rather than of focusing on a single critical episode; this approach allows to better evaluate the model performances and to quantify policies effects with respect to long-term air quality standards. Several ozone modelling studies were already

performed for Portugal, in β -mesoscale domains and during some specific and episode days (e.g., Barros et al., 2003; Borrego et al., 2000). Large-scale simulations including Portugal have also been done, but with a coarse grid resolution (Coutinho and Borrego, 1991). The present work represents an important and a pioneer scientific air pollution modelling study for Portugal, because it aims to assess the air quality over the whole continental region of Portugal for the most critical gaseous pollutants (SO_2 , CO, NO_2 and O_3) and during the entire year of 2001, using a modelling system that was evaluated taking into account the EC Modelling Quality Objectives.

2. Methodology

CHIMERE is a 3D chemistry-transport model, based on the integration of the continuity equation for the concentrations of several chemical species in each cell of a given grid (Schmidt et al., 2001). CHIMERE has been used for several research applications, including sensitivity to anthropogenic or biogenic emissions (e.g., Beekmann and Derognat, 2003; Menut, 2003; Derognat et al., 2003), emission diagnostics (Vautard et al., 2003), or photo-oxidant forecasting over several regions of Europe (namely Italy and Belgium), being the official model for air quality forecasting over France and Paris region (Vautard et al., 2000). Results showed a reasonable skill for daily maximum forecasts of ozone with an averaged root mean square (RMS) error of about 10 ppb and 0.8 of correlation, which is in agreement with the ozone forecast model intercomparison experiment described in Tilmes et al. (2002). Besides this

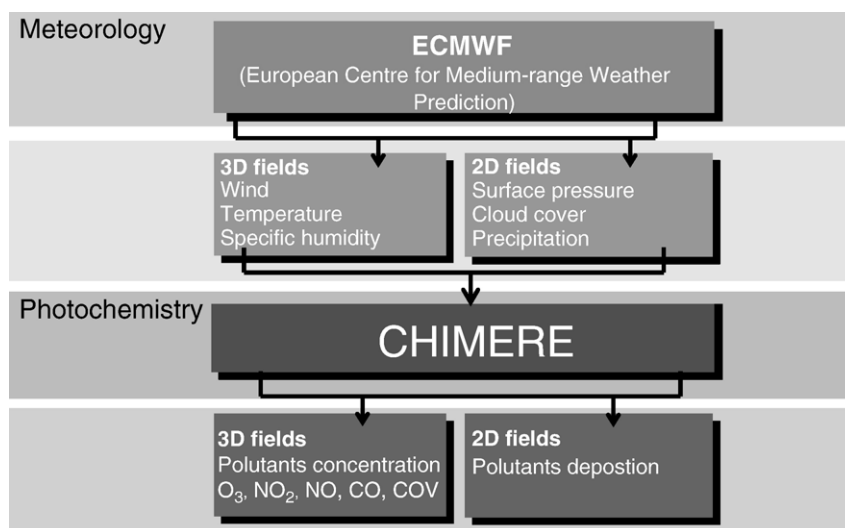


Fig. 1. Structure of the modelling system.

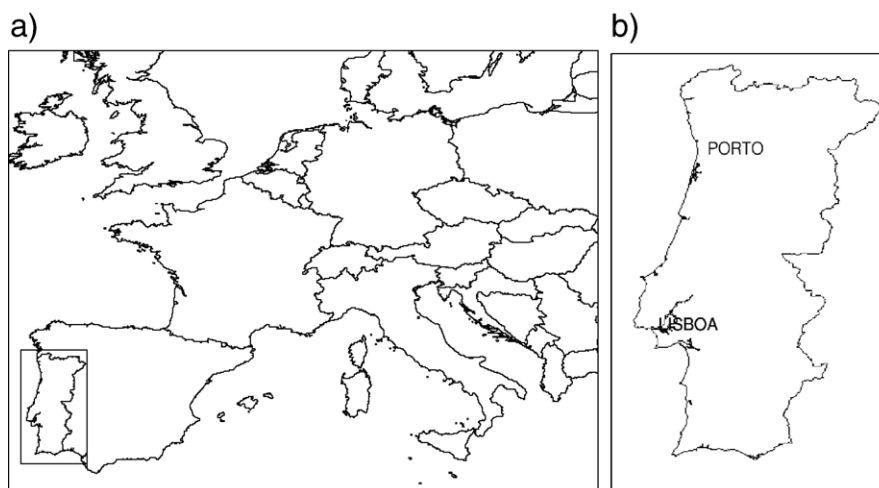


Fig. 2. Geographical domains used by the CHIMERE model.

proposed assessment task, this modelling system is being also applied in real-time air quality forecasting over Portugal (Monteiro et al., 2005a). The model version used here is primarily described in Schmidt et al. (2001), and further updates, especially for the smaller-scale version, can be found in Vautard et al. (2003). The meteorological input variables are taken, as in Schmidt et al. (2001), from the European Centre for Medium-range Weather Forecast (ECMWF): 3D fields of horizontal wind, temperature, specific humidity, cloud liquid water content, and 2D fields of surface pressure, heat fluxes, 2 m temperature and cloud cover, with different resolutions according to the domain of simulation (36×36 km for the European domain and 9×9 km resolution for the Portugal domain). They are linearly interpolated to the CHIMERE grid, and linear time interpolation is also applied to obtain hourly values, along 2001. Besides the meteorological input, the CHIMERE model needs boundary and initial conditions, emission data, and the land use and topography characterization. The choice of year 2001 was conditioned by the last revised national emission inventory availability. In addition, CHIMERE simulations do not yet include forest fires emissions; therefore 2001 was considered a reliable period to simulate because it was not a critical year in terms of forest fires. Fig. 1 presents a simplified scheme of the CHIMERE model and its inputs/outputs.

The model was applied first to a continental scale (from 10.5°W to 22.5°E and 35°N to 57.5°N , see Fig. 2a) and then to Portugal, as shown in Fig. 2b, using the same physics and a simple one-way technique. The second simulation (Portugal domain) was performed with a horizontal domain of $290 \text{ km} \times 580 \text{ km}$ and a

10 km horizontal resolution (using the ECMWF data with 9×9 km resolution) and the vertical grid consisted of six hybrid sigma-pressure layers with a model top at 700 hPa. The top altitudes of the layers vary with time, but their approximate values are, from bottom to top: 50, 250, 600, 1200, 2000, and 3000 m.

Lateral and top boundaries for the large-scale run are obtained by the MOZART second-generation model monthly climatology (Horowitz et al., 2005) as described in Monteiro et al. (2005b). As regards emissions, the CHIMERE model requires the input of 15 primary compounds: NO, NO_2 , HONO, SO_2 , CO, ethane, *n*-butane,

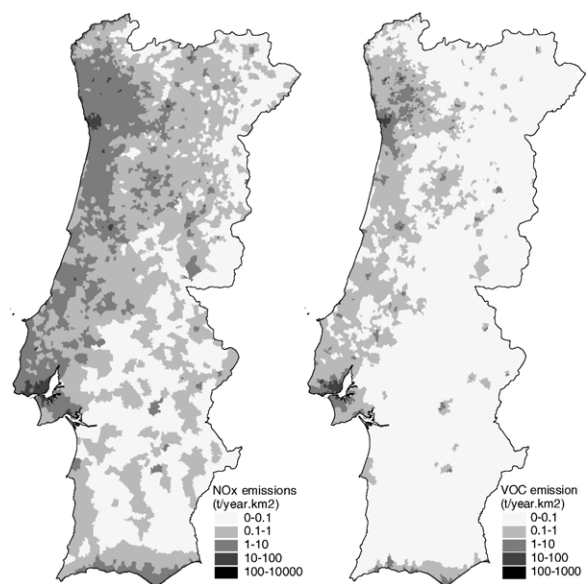


Fig. 3. Spatial distribution of NOx and VOC emissions from area sources.

Table 1

Limit and target values defined by the European Union directives for SO₂, NO₂, O₃, and CO

Parameters	Period	Air quality standards ($\mu\text{g m}^{-3}$)	No. of allowed exceedances	Target date
<i>SO₂ (1999/30/EC)</i>				
Human health protection	Hourly average	350	24 times/year	1 Jan 2005
Human health protection	Daily average	125	3 times/year	1 Jan 2005
Vegetation protection	Annual average	20	—	—
Vegetation protection	Winter average	20	—	—
<i>NO₂ (1999/30/EC)</i>				
Human health protection	Hourly average	200	18 times/year	1 Jan 2010
Human health protection	Annual average	40	—	1 Jan 2010
<i>CO (1999/30/EC)</i>				
Human health protection	8 h running average	10 mg m ⁻³	—	1 Jan 2005
<i>O₃ (1999/30/EC)</i>				
Human health protection (target value)	8 h running average	120	25 times/year	1 Jan 2010
Vegetation protection (AOT40 target value)	Hourly average (May–July)	18 000 $\mu\text{g m}^{-3} \text{ h}$	—	1 Jan 2010
Information threshold	Hourly average	180	—	—
Alert threshold	Hourly average	240	3 consecutive h	—

ethene, propene, isoprene, α -pinene, *o*-xylene, formaldehyde, acetaldehyde, and methyl ethyl ketone. For European scale, emissions were derived from the annual totals of the EMEP database for 1999 through a methodology similar to that described in Schmidt et al. (2001).

Over the Portuguese domain, area source (namely transport, combustion and industrial processes, waste treatment, and agriculture) data were obtained following different methodologies as described in Monteiro et al. (2005b). Examples of the estimation of spatial distribution of emissions [NO_x and volatile organic compounds — (VOC)] from this approach are shown in Fig. 3. Both maps show that most of the emissions are concentrated along the western coast of Portugal, where the two main urban centers of Lisbon and Porto are located. The traffic sector represents, compared to industrial activities, a major source of air pollution (Borrego et al., 2002).

Large point source annual emissions were obtained directly from the available monitoring data of each industrial plant. Time disaggregation was calculated by application of monthly, weekly, and hourly profiles obtained in the scope of GENEMIS Project (GENEMIS, 1994). The non-methane volatile organic compounds (NMVOCs) are disaggregated into 227 individual VOCs according to the speciation suggested by Passant (2002) for each activity sector referred above. The methodology for biogenic emissions of isoprene and terpenes is described in Schmidt et al. (2001). The land use database comes from the Global Land Cover Facility (Hansen et al., 2000), which, after some processing, provides the cell-by-

cell coverage of coniferous and broadleaf forests. The spatial distribution of tree species within these classes is established following the methodology outlined in Simpson et al. (1999). The Stohl et al. (1996) methodology is used for biogenic emissions of NO from fertilized soils.

3. Model validation

The FWD and daughter directives establish requirements for air quality modelling, including the definition of the Modelling Quality Objectives, as a measure of acceptability of modelling results. In this context, the uncertainty for modelling and objective estimation is defined as the maximum deviation of the measured and calculated concentration levels, over the period for calculating the appropriate threshold, without taking into account the timing of the events. The quality objectives defined for each quality indicator are listed in Table 1.

The model quality measure described in the EU directives is interpreted as the maximum error without timing. For the cases where the absolute maximum error is calculated at the highest percentile, that is, at the highest measured value, the assessment of the model accuracy depends on the model performance in a concentration range associated with an extremely small probability. This also means that the model accuracy assessment can be based on an outlier concentration, caused by an error of the monitoring unit or an extreme weather situation. Therefore, an alternative model error measure is proposed by Stern and Flemming (2004), which is defined as the

Table 2

Relative error estimation for each pollutant legislative percentile, considering an average of all the monitoring stations

Pollutant	Legislated indicators	Relative error (%)
SO ₂ (EU directive 1999/30/EC)	Human health protection (25th maximum hourly average)	34
	Human health protection (fourth maximum daily average)	57
	Vegetation protection (annual average)	43
	Vegetation protection (winter average)	54
NO ₂ (EU directive 1999/30/EC)	Human health protection (19th maximum hourly average)	48
	Human health protection (annual average)	50
CO (EU directive 2000/69/EC)	Human health protection (8 h running average)	32
O ₃ (EU directive 2002/3/EC)	Human health protection (26th maximum 8 h daily average)	16
	Vegetation protection (AOT40)	49

concentration difference at the percentile threshold corresponding to the allowed number of exceedence of the limit value normalized by the observation. This relative error (rel_per_err) can be defined by the following expression, where Op and Sp represent the observations and calculations, respectively, for each percentile threshold legislated:

$$\text{rel_per_err} = \frac{|\text{Op} - \text{Sp}|}{\text{Op}}$$

In this way, this measure is more robust than the error defined in the EU directive and also evaluates the model performance in the high-concentration ranges, but without the sensitivity to outliers. Following these data quality objectives, a relative error at each legislated percentile threshold was calculated for each air quality background-monitoring site from the national air quality-monitoring network (a total of 15 stations located mainly near the urban centres of Porto and Lisbon), to evaluate model performance for the main critical pollutants. Traffic stations were excluded from this statistical analysis because the model resolution used in this application ($10 \times 10 \text{ km}^2$) is not representative for those monitoring sites. Table 2 presents the relative error average for all the monitoring sites, for each pollutant parameter defined by the respective daughter directive.

The analysis of Table 2 shows that the model system is able to reproduce the various legislated percentiles, because the relative error measure complies with the accuracy requirement of 50% for almost all the pol-

lutants' limit thresholds. An analysis by pollutant species indicates that ozone presents lower deviations. This fact could be expected because ozone is a secondary pollutant, not directly influenced by emissions as the NO₂ pollutant. Besides that, the gas-phase version of the CHIMERE model applied in this study (the aerosol version is still under testing for Portugal) was specially developed to simulate oxidant pollutants and photochemistry and did not include the aerosols' chemistry, which could justify the noncompliance of some parameters for the SO₂ pollutant.

Besides this quantitative model evaluation, a qualitative comparison between the modelling results and the average concentrations of NO₂ and O₃ obtained in a diffusive sampling experimental campaign that took place during two weeks of 2001 (7–21 May and 21–29 June) was performed. This sampling campaign (Ferreira et al., 2001) covered the entire Portuguese continental territory with a sampling diffusive points distribution, within a spatial grid of $20 \times 20 \text{ km}^2$ resolution, enabling a broader spatial distribution of NO₂ and O₃ over Portugal than the one represented by the national air quality network. The measured values represent weekly averaged concentrations. Fig. 4 presents the comparison between the observed and modelled (at first vertical level) NO₂ averaged values over the first sampling week and their spatial distribution, showing a relatively good correspondence and a satisfactory model performance. Nevertheless, the model overestimates the NO₂ average concentrations in specific areas. In fact, some of these sites correspond to large point source locations, suggesting that the emissions from these industrial sources were introduced in a lower layer of the model, neglecting the plume rise effect. Also, the modelled average of the metropolitan areas of Lisbon and Porto is higher than the observed. This could be justified by an overestimation of the emission inventory for these particular areas (Borrego et al., 2002).

Regarding ozone, the average concentration maps show some discrepancies between the experimental campaign data and the modelling results (Fig. 5), especially in areas not covered by the national monitoring network, such as the north and southeast region. In fact, taking into account that the wind direction during this specific period had a strong east component, in opposition to the N–NW dominant wind direction, this could be due to a lack of knowledge about the emission inventory of Spain (namely point sources emissions) and a consequent underestimation of the boundary conditions in that part of the domain. This hypothesis is under investigation through a scientific collaboration established between research groups of Spain (Galicia) and Portugal.

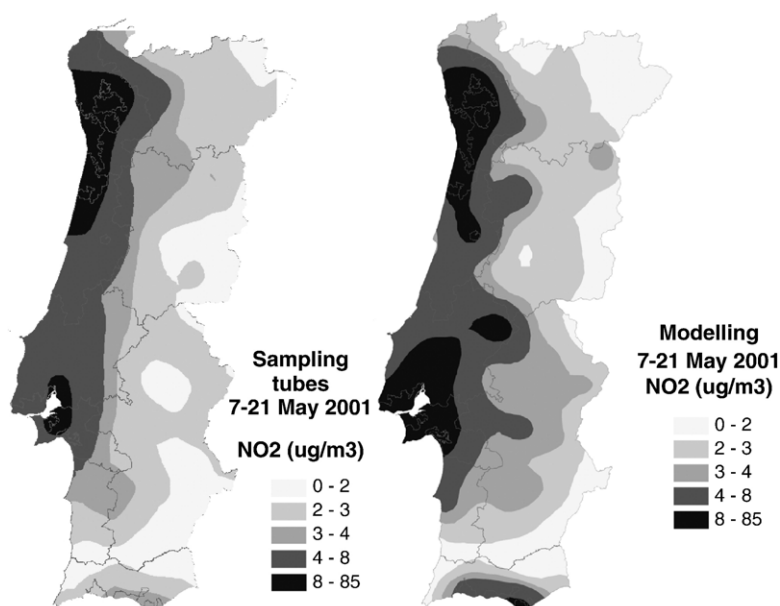


Fig. 4. Weekly averaged concentration maps of NO_2 resulting from sampling tubes campaign (Ferreira et al., 2001) and numerical modelling simulations during the period of 7–21 May 2001.

Nevertheless, it remains a good model performance for the major part of Portugal, especially where the monitoring stations from the national network are mainly located, confirmed by the statistical validation presented in Monteiro et al. (2005b) with correlation coefficients of 0.7 and errors less than $30 \mu\text{g m}^{-3}$ for ozone.

4. Results and discussion

The model validation exercise for 2001 initiated in Monteiro et al. (2005b) and developed in the present work shows a model system able to simulate the gaseous pollutants and to perform the first air quality assessment

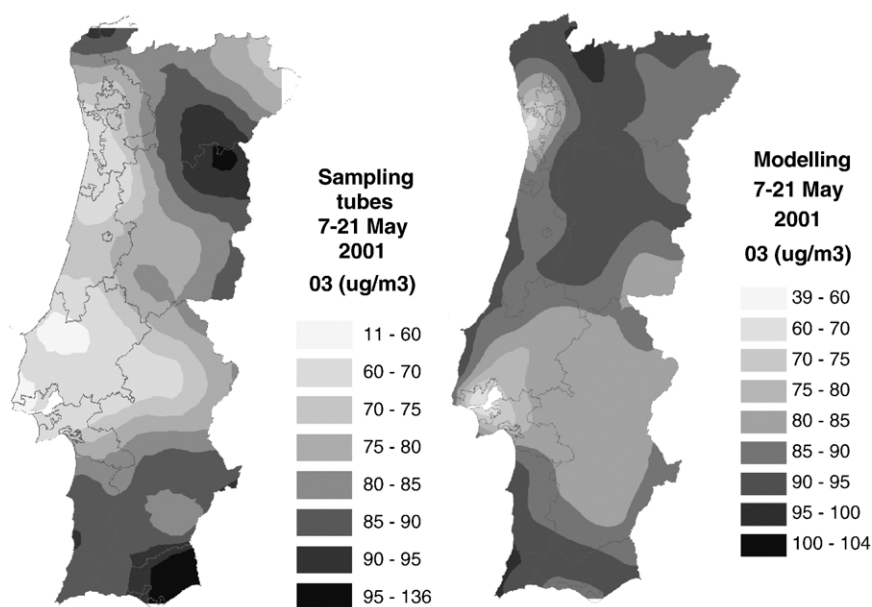


Fig. 5. Weekly averaged concentration maps of O_3 resulting from sampling tubes campaign (Ferreira et al., 2001) and numerical modelling simulations during the period of 7–21 May 2001.

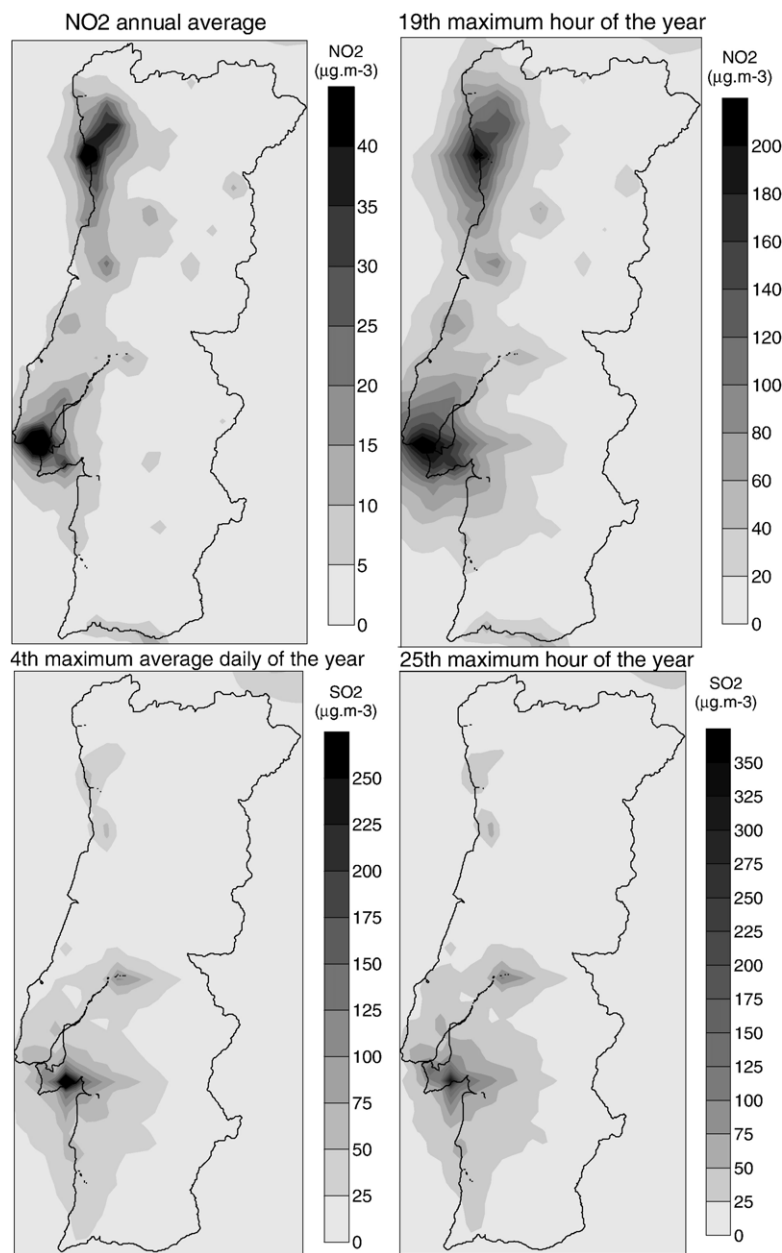


Fig. 6. Modelling results for NO₂ and SO₂ considering human health protection limits for the year 2001.

for Portugal. The modelling results for 2001 were analyzed according to the limit and target values defined by the EU directives for each pollutant considered in this study (namely SO₂, NO₂, CO, and O₃). Fig. 6 presents the modelling results, for 2001, for the legislation indicators of human health protection regarding NO₂ (annual average and 19th maximum hour of the year) and SO₂ (fourth maximum daily average and the 25th maximum hourly average).

With regard to NO₂, there are areas presenting values exceeding the limit parameters, which correspond to the main urban areas of Portugal: Porto and Lisbon metropolitan regions. Because the remaining area does not present any critical values, this suggests that road traffic is the main activity responsible for the above-mentioned high concentrations of NO₂. The modelling results related to SO₂ show that there are two areas where the concentrations exceeded the limit values, identified as

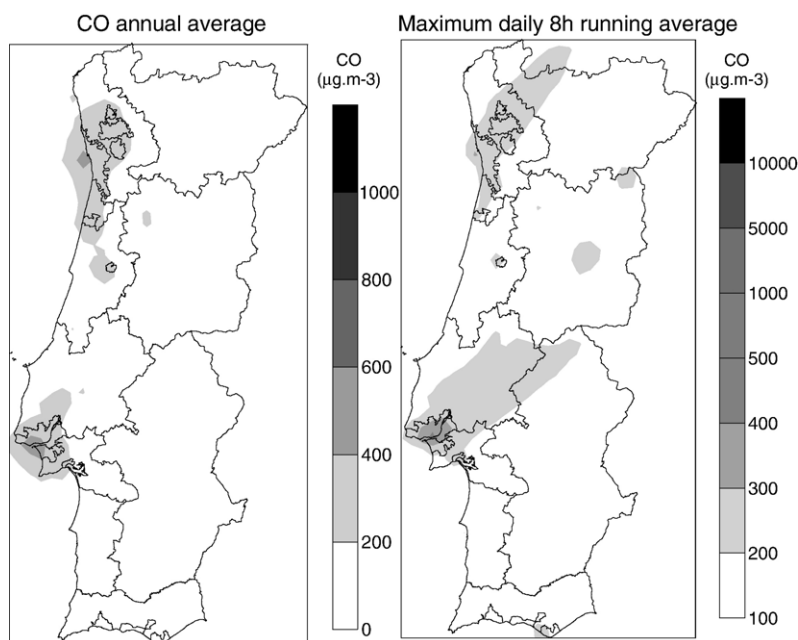


Fig. 7. Modelling results for the annual average and maximum daily 8 h running average (human health protection limit) of CO for the year 2001.

industrial places, which suggests that the industry activity is the main factor responsible for the bad air quality in these areas.

For CO, the legislation establishes a maximum of 10 mg m^{-3} for the daily 8 h running averages for human health protection. Fig. 7 presents the CO annual average

and the daily 8 h average concentration patterns. It is possible to verify that there are no areas with concentrations higher than this limit. In fact, the simulation results indicate very low annual average concentrations ($200\text{--}1000 \text{ µg m}^{-3}$) for this pollutant for the entire region of Portugal. This was already expected and

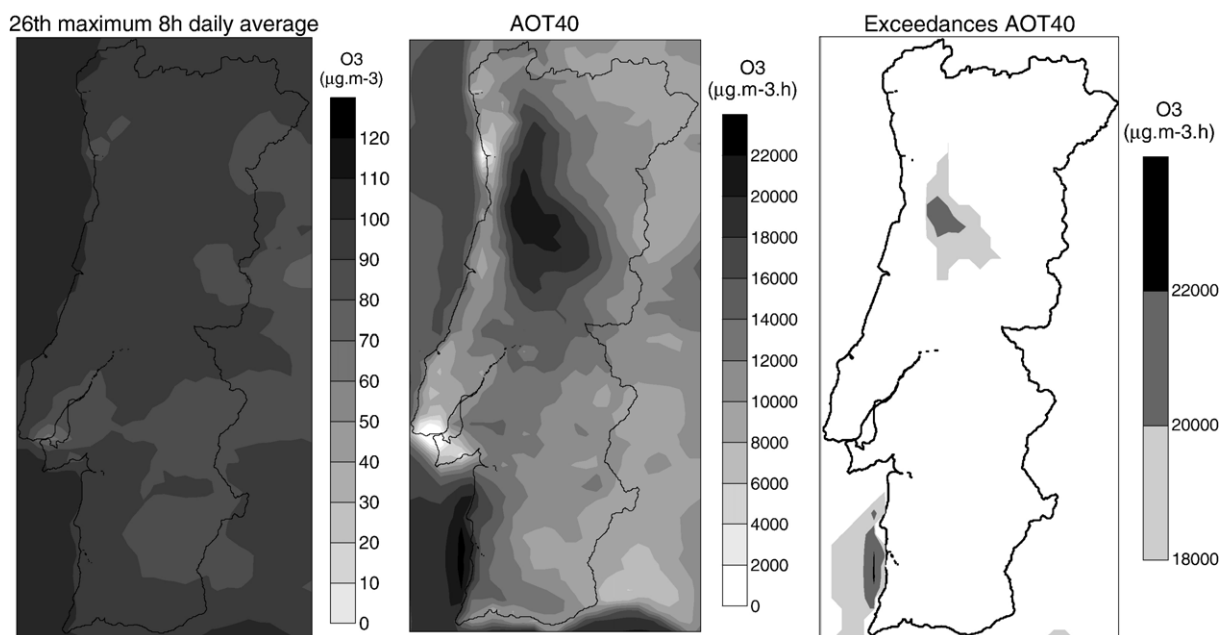


Fig. 8. Modelling results for O₃ human health and vegetation protection limits for the year 2001.

confirms the modelling and experimental studies that point out CO concentrations of 0.2 mg m^{-3} (150 ppb) in remote sites of Europe and values of $1\text{--}10 \text{ mg m}^{-3}$ in urban areas (Pfister et al., 2004).

The third daughter directive (2002/03/EC), concerning ozone, defines the target value of $120 \text{ } \mu\text{g m}^{-3}$ for the 26th maximum 8 h daily average and a maximum of $18,000 \text{ } \mu\text{g m}^{-3} \text{ h}$ for the AOT40 value. Fig. 8 presents the modelling ozone concentration fields for these two parameters.

Despite the fulfilment of the target value for human health protection ($120 \text{ } \mu\text{g m}^{-3}$) for the entire national territory, there is some risk for vegetation protection in areas with values exceeding the AOT40 threshold. Unfortunately, according to the land use map, these areas correspond to high-density vegetation and forest areas. These ozone concentration patterns were already foreseen in previous mesoscale modelling studies (Barros et al., 2003; Borrego et al., 2000), where the ozone precursors transport from the urban areas of Porto and Lisbon to the south and interior part of Portugal (by the N–NW dominant winds) is clear and significant.

5. Conclusions

This work aimed to assess air quality in the continental part of Portugal, according to the requirements of the new FWD air quality. This first assessment exercise was made for the year 2001, using the CHIMERE chemistry-transport model. The model system was evaluated by comparison with observational data obtained from the regional air quality monitoring networks for the main critical gaseous pollutants (NO_2 , SO_2 , CO, and O_3). The estimated errors in this validation exercise are relatively small and below the limits established by legislation for model evaluation.

With regard to the fulfilment of the thresholds defined by each daughter directive, the modelling results show that there are values exceeding the human health protection limit for NO_2 in the two main urban areas (Porto and Lisbon), probably due to road traffic emissions. In comparison, for SO_2 , the concentration fields indicate some small areas with higher values (higher than the human health limit), all of them located close to large point sources (power plants, more precisely). Ozone is another pollutant with areas exceeding the legislated target values, in what concerns vegetation protection. These areas are mainly located in the inland central part and southwest coast of Portugal, not covered by the air quality monitoring networks. CO is the unique pollutant that accomplishes the limits imposed by legislation, with concentration values significantly low in the entire study domain.

This first air quality assessment work for Portugal enabled to verify the importance of the establishment of several target values for each air pollutant. In fact, the fulfilment of one of these limits does not imply the accomplishment of all, as confirmed by the O_3 results. Future work will focus on the other critical pollutants, namely PM_{10} , and will evaluate the CHIMERE aerosols version for this specific coastal area. Besides that, simulations on urban scale for Lisbon and Porto domains will be performed, with a higher-scale resolution ($2 \times 2 \text{ km}^2$), to evaluate more correctly the air quality problems in these regions, identified as the main critical areas, and influenced by traffic emissions and local conditions.

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